学 位 論 文 審 査 の 要 旨
Doctoral Dissertation Evaluation Review

博士の専攻分野の名称　博士（生命科学）　　氏　名

Degree requested: Doctor of （Life Science）　　Name: CHANCHAL KUMAR ROY

審査担当者	主査 / Chief examiner	教　　授	龔　剣萍
Examiners	副査 / Associate examiner	准 教 授	黒川　孝幸
	副査 / Associate examiner	特任教授	佐々木　直樹

学 位 論 文 題 名
Title of Doctoral Dissertation

Adhesion of Polyampholyte Hydrogels
（ポリアンフォライトハイドロゲルの接着）

博士学位論文審査等の結果について（報告）
Results of Evaluation of the Doctoral Dissertation (Report)

Hydrogels are one of the most attractive biomaterials in life science. They have wide possible applications ranging from drug delivery to tissue engineering. For most of the applications, bonding of tissue and hydrogel is necessary. However, most hydrogels have poor interfacial adhesion with different substrates in wet environment, which have greatly limited their practical applications. The author has proposed that the adhesion problem of hydrogels can be solved by using polyampholyte (PA) based hydrogel system. PA hydrogels are newly developed hydrogels having dynamic ion-bonds in their bulk material and surface. The ionic bonds of PA chains at surfaces can switch their ion-pair for the formation of attractive interaction sites at interface. Using the advantage of dynamic ionic bonding PA hydrogel can make strong adhesion on various substrates ranging from soft and wet biotissue to solid substrates in wet environment. PA hydrogel seems to be an interesting material for adhesive use. In this dissertation the author has investigated the adhesion behavior of PA hydrogels in wet environment over different substrates. Here we are reporting the results of evaluation of the doctoral dissertation.

This doctoral dissertation consists of six chapters. The extent of this thesis corresponds its significance and amount of new knowledge it presents. The problem of poor adhesion of common hydrogels and their possible solving strategies have been systematically discussed in Chapter 1. A detailed review of polyampholyte systems has been carefully organized in Chapter 2. In Chapter 3 a new adhesion phenomena has been explored for hydrogels. It has been demonstrated that a typical PA hydrogel, (P(NaSS-*co*-DMAEA-Q)) prepared from their monomers; sodium 4-vinylbenzenesulfonate (NaSS) and (2-acryloyloxyethyl)trimethylammonium chloride quaternary (DMAEA-Q), can make strong adheison with tissue materials. To explore the mechanism of strong adhesion, the author has investigated the adhesion behavior of PA hydrogel over charged and uncharged hydrogel substrates using lap shear test. Interestingly, it has been observed that PA hydrogel can make strong adhesion with both positively charged and negatively charged hydrogels. This special kind of adhesive property of PA hydrogel has been named as self-adjustable adhesion of PA hydrogel. The author has introduced self-adjustable adhesion phenomena of a hydrogel for the first time. It has been reported here that through the self-adjustable adhesion meachanism, PA hydrogel can make large number of adhesive interacting sites over tissue surfaces which is responsible for their strong adhesion over tissue surfaces. The author has used the findings of this section for publishing an article in interantional journal as a first author (Self-Adjustable Adhesion of Polyampholyte Hydrogels, *Advanced Materials*, **2015**, *27*, 7344-7348).

In this chapter, the author has tried to determine the critical energy release rate (G_c, geometry independent parameter of adhesives) of PA hydrogel over charged hydrogels, but failed due to the weak mechanical property of polyelectrolyte gels. However, the author has inteligently overcome this problem by using negatively charged glass substrate. Chapter 4 deals with the determination of G_c of PA hydrogel over solid substrates in wet environment. The estimated G_c, ~116 J/m^2 over glass plate is much higher than the adhesion energy of PDMS elastomers that is in the range of 10-30 J/m^2. The large value of adhesion energy has been related to the strong adhesive interaction at interface and additional viscoelastic energy dissipation in the bulk material. Strong interfacial contact formation and viscoelastic energy dissipation occur in PA hydrogel adhesive due to the presence of dynamic ionic bonding of monomeric units inside the material. In Chapter 5, the adhesion behavior of PA hydrogel (P(NaSS-*co*-DMAEA-Q)) has been systematically investigated over glass plate at different conditions. The preparation conditions of PA hydrogel have been explained for achieving maximum adhesion. The author has observed that the composition of the PA hydrogel and dialysis time plays an important role in its adhesive behavior. A neutral charged balanced PA hydrogel has better adhesion over glass substrate than charged imbalanced hydrogels. PA works well for hydrophilic substrates than hydrophobic substrates. Underwater tack test has been performed to investigate the adhesion behavior of PA hydrogel over a wide range of pH and temperature. Although more details explanation about the parameters influencing the adhesion behavior of PA hydrogel was necessary but we think that the author can leave this study for future work. Finally, in Chapter 6, the author has proposed a strategy to develop sticky and tough supramolecular hydrogels. The idea is to use polymer that form abundant of dynamic bonds, such as ionic bond, hydrogen bond, etc. The proposal of this idea will certainly open a new avenue in the research of development of adhesive hydrogels.

After careful examination of this thesis we are convinced that it presents significant and original contribution to the scientific field it deals with. The findings and understanding of this dissertation can be used not only in the field of life science but also it can contribute to the different branches of material science especially in adhesive science. The thesis fulfills all requirements for obtaining the academic degree of Doctor of Life Science. Therefore, we acknowledge that the author is qualified to be granted the Doctorate of Life Science from Hokkaido University.